



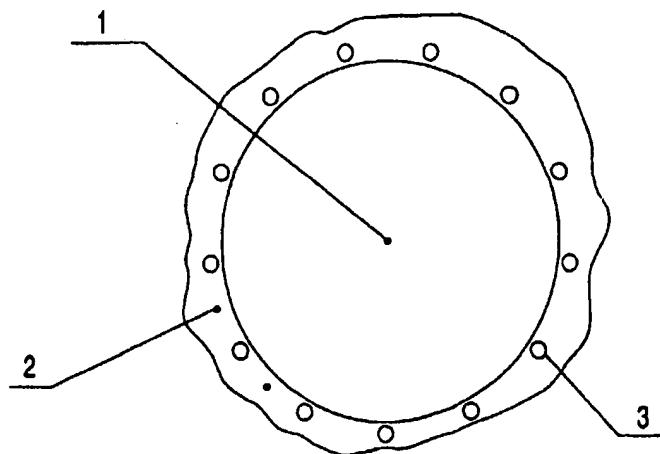
## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

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(54) Title: NEW GLASS/POLYMER COMPOSITES

## (57) Abstract

A glass/polymer composite material including an organic polymer (1) and chalcogenide glass (3) and process of manufacturing same.



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## NEW GLASS/POLYMER COMPOSITES

FIELD AND BACKGROUND OF THE INVENTION

The present invention relates to polymeric composite materials including organic polymers characterized by having a low melting point and inorganic chalcogenide glasses, and to a process for their preparation.

The advantages of reinforcing a thermoplastic polymer with a filler such as an inorganic glass are well documented in the literature. Polymer/glass composites are known to exhibit superior characteristics compared to non-composite polymer or glass taken individually. Improved characteristics include, *inter alia*, increased thermal stability, chemical stability, and enhanced fracture toughness. This unique combination of characteristics has led to the use of polymer/glass composites in a wide range of applications, such as fire retardants, optical windows, protective coatings and packaging materials with barrier properties. The thermoplastic polymer/glass materials can take the form of a barrier coating, a flexible film or a rigid film. Such a film sheet or coating material can act as a barrier to liquids, water vapor and other gases.

The polymer/glass composites are made in several forms. U.S. Patent 5,043,369 describes a method in which the glass (and/or glass-ceramic) and thermoplastic and/or thermosetting polymer(s) are combined in a melt form at compatible working temperature and viscosity to form an intimate mixture. In other words, the glass and polymer are in a sufficiently fluid state to be blended together to yield a uniform, fine grained microstructure body. Such microstructure organization may be (a) fine, spherical, ellipsoidal and/or serpentine particles of glass and/or glass-ceramic separated by thin membranes of polymer; (b) an interlocking, 3-D microstructure comprising islands of glass and/or glass-ceramics in polymer; (c) a fine emulsion of glass and/or glass-ceramic dispersed in polymer; and (d) a fine emulsion of polymer dispersed in glass and/or glass-ceramic. Mixtures of the glass and the polymer are made in two major forms: a glass-phase dispersed constituent within the polymer matrix phase or a polymer-phase dispersed material distributed throughout the glass matrix phase. In either form, the microstructure of the polymer and/or the glass constituents persists as the phases are mixed. U.S. patent 5,422,384 discloses a method for preparing a polymer/glass composites wherein the interfacing between the polymer and glass constituents or phases is at the molecular level thereby making the polymer and the glass constituents

practically indistinguishable. The method of making these molecular phase polymer/glass composite materials includes the steps of using a compatible polar solvent in which the solid glass and the polymer are dissolved to form a homogeneous mixture of the solutes at the molecular level; removing the solvent and heating the homogeneous molecular mixture to form bonds at the molecular level between the glass and the polymer.

U.S. Patent 3,732,181 discloses composite materials containing low melting inorganic oxide glass, preferably phosphorus and/or boron oxides of which lead and zinc phosphate glass are preferred, and a wide variety of organic thermoplastic polymers, preferably high density polyethylene, polypropylene, poly-4-methylpentene-1, polyethylene terephthalate, polysulphones, polycarbonates, polytetrafluoroethylene, polyvinyl chloride and polystyrene. The inorganic oxide glass has a transformation temperature ( $T_g$ ) in the range of 100-400°C and it softens between the softening temperature of the thermoplastic polymer and the highest temperature at which the polymer is chemically stable.

U.S. Patent 5,043,369 reports on polymer/glass composites containing phosphate-based glasses having low transition temperature. The patent points out three essential characteristics of the glasses to be used in the polymer/glass composite: (1) a sufficiently low  $T_g$  to be thermally co-deformable with organic polymers; (2) the capability of interacting with a polymer so as to form bodies having essentially uniform, fine-grained microstructures; and (3) exhibit excellent resistance to attack by moisture, i.e., the glass must be non-hygroscopic.

U.S. Patent 4,141,877 discloses polymer/glass composites composed of hydrated alkali metal silicate glass and an organic polymer resistant to a hot, aqueous and alkaline environment.

Many of the above described processes for preparing polymer/glass composites deal with a relatively high working temperature due to a relatively high softening temperature point of the glass used. This limitation was stated in the U.S. Patent 3,732,181: "Not all polymers are co-deformable with all glasses. For example polyethylene has too low a melt-viscosity to be used in certain composites forming processes with glasses having a transformation temperature in the upper part of the range". Although, this U.S. patent suggests that HD polyethylene can be formed into composites using some of the lower softening point inorganic oxide glasses, it seem inevitable that a better solution should be found for polymers having low- softening and melting temperature points.

The use of linear triblock copolymer elastomer compounds is taught by U.S. Patents 5,187,236 and 5,210,359. These elastomers are herein referred to as RMR liquid elastomers. These two U.S. Patents are incorporated by reference for all purposes as if fully set forth herein. These RMR-elastomers are liquid triblock copolymers with a narrow molecular weight distribution. The center block is a random ethylene/butene backbone. The terminal blocks contain carbon-carbon double bonds for curing with sulfur or peroxide.

Further, the prior art teaches methods of preparing bulk phase polymer/glass composites (in which the microstructure of either or both phases persists as the phases are mixed) based on either thermo-mechanical bulk mixing or diffusion processes. Generally speaking, the glass constituent and/or the polymer constituent are heated and softened at a compatible working temperature, or melted at an elevated temperature (approximately 400 degrees C) while mechanically mixed. Neither of the methods described teaches a procedure in which the glass particles are selected from the group of chalcogenide compounds nor a procedure in which the glass particles are directed to adhere as a distinct separate layer on the surface of the polymer particles using a particular elastomer material which glues the glass particles to the surface of the polymer particles.

The chalcogenide glasses are characterized by combinations of elements mainly selected from the semi-metal group. Examples of typical chalcogenide glasses are combinations of arsenic (or arsenic-antimony) with sulfur or selenium or a combination of sulfur and selenium. It is possible to add additional elements such as iodine, tellurium and germanium. Chalcogenide glasses include no oxygen and are further characterized by having low softening temperature points (90-200° C) and low viscosity (10-1000 PaS). These properties make the chalcogenide glasses very suitable for being used in composites with polymers having low softening and melting points.

In addition, chalcogenide glasses have some unique optical properties: a) these glasses are transparent in the middle and far IR bands of the spectrum ( in the interval: 2-20 mm); b) there is no dispersion in the cited above band; c) they have extremely high refractive index (up to 2.5); and d) they are characterized by photorefractive effect.

There is thus a widely recognized need for, and it would be highly advantageous to have a glass/polymer composite with low softening

temperature and low viscosity which is suitable for use in applications including, but not limited to, infrared optics.

#### SUMMARY OF THE INVENTION

According to one aspect of the present invention there is provided a glass/polymer composite material including an organic polymer and chalcogenide glass.

According to further features in preferred embodiments of the invention described below, the organic polymer constituent of the glass/polymer composite material has a low melting point.

According to still further features in the described preferred embodiments, the glass and polymer constituents of the glass/polymer composite material have the same or similar softening temperature points in the range of 90-200°C.

According to still further features in the described preferred embodiments, the glass and polymer constituents of the glass/polymer composite material have the same or similar softening temperature points in the range of 90-200°C and the same or similar viscosity values in the range of 10-1000 PaS under extrusion conditions.

According to still further features in the described preferred embodiments, the glass/polymer contains low density polyethylene and chalcogenide glass consisting of As - 15%, Sb - 3%, I - 28%, Se - 50%, Te - 4% ( $\text{As}_{15}\text{Sb}_3\text{I}_{28}\text{Se}_{50}\text{Te}_4$ ).

According to another aspect of the present invention there is provided a glass/polymer composite material including: an organic polymer, a glass and a linear triblock copolymer elastomer.

According to still further features in the described preferred embodiments, the glass/polymer composite material includes glass selected from the group consisting of chalcogenide and alkali metal based glasses.

According to still further features in the described preferred embodiments, the glass/polymer composite material includes an elastomer which is a linear triblock copolymer having a narrow molecular weight distribution and having a center block which is a random ethene/butene backbone and terminal blocks which contain carbon-carbon double bond.

According to still further features in the described preferred embodiments, the glass/polymer composite material includes polymer which constitutes from 90 to 99.5% by weight of the material, and glass which constitutes from 0.5 to 10% by weight of the material. According to still

further features in the described preferred embodiments, the glass/polymer composite material contains 99% by weight polymer and 1% by weight glass.

According to still further features in the described preferred embodiments the glass/polymer composite material is designed useful as a light filter, an optical window, a polarization layer, an adhesive and as a light shaper/diffuser.

According to yet another aspect of the present invention there is provided a process for preparing a glass/polymer composite containing organic polymer particles and chalcogenide glass particles including the steps of: (a) mechanically milling of the glass particles to a size of  $< 100 \mu\text{m}$ ; (b) mixing the milled glass particles with the polymer particles; and (c) extruding the glass and polymer particles simultaneously in a melted form.

According to still another aspect of the present invention there is provided a process for producing a glass/polymer composite material including particles of an organic polymer, glass particles and linear triblock copolymer elastomer, including the steps of: (a) mechanically milling the glass particles to a final size of  $< 100 \mu\text{m}$ ; (b) homogeneously dispersing the milled glass particles in a solution of the elastomer in a solvent; (c) adding the polymer particles into the mixture of glass particles dispersed in the elastomer solution; (d) evaporating the solvent to form polymer particles coated by a layer of elastomer in which the glass particles are dispersed; and (e) extruding the glass and coated polymer particles simultaneously in a melted form.

According to still further features in the described preferred embodiments, the extrusion step of the process occurs under conditions of high shear stresses ( $10^5 - 10^6 \text{ Pa}$ ).

According to still further features in the described preferred embodiments, the process results in production of a film of 20-100  $\mu\text{m}$  thickness.

The present invention successfully addresses the shortcomings of the presently known configurations by providing glass/plastic composites with low glass content and processes for forming same at relatively low temperatures and under conditions of high shear stress.

#### BRIEF DESCRIPTION OF THE DRAWINGS

The invention is herein described, by way of example only, with reference to the accompanying drawings, wherein the sole figure shows a

polymer particle having on its surface a distinct layer of elastomer in which glass particles are dispersed.

#### DESCRIPTION OF THE PREFERRED EMBODIMENTS

The present invention is of polymeric composite materials including organic polymers characterized by having a low melting point and inorganic chalcogenide glasses, and to processes for their preparation.

As used herein in the specification and in the claims section that follows, the phrase "composite material", includes any stable mixture of the polymer(s) and glass. Thus, for example, homogenous, layered and emulsion mixtures, examples thereof are provided in the background section above, are all included under the definition of this phrase.

The polymeric composite materials are suitable for use in applications including, but not limited to, infrared optics.

Specifically, the present invention can be used in a wide range of applications, such as fire retardants, optical windows, protective coatings and packaging materials with barrier properties. The thermoplastic polymer/glass materials can take the form of a barrier coating, a flexible film or a rigid film. Such a film sheet or coating material can act as a barrier to liquids, water vapor and other gases

The principles and use of a polymeric composite material and processes for preparation of same according to the present invention may be better understood with reference to the drawing and accompanying descriptions and examples.

Before explaining at least one embodiment of the invention in detail, it is to be understood that the invention is not limited in its application to the details of construction and the arrangement of the components set forth in the following description or illustrated in the drawings. The invention is capable of other embodiments or of being practiced or carried out in various ways. Similarly, the invention may be used for purposes not specifically set forth hereinbelow. Also, it is to be understood that the phraseology and terminology employed herein is for the purpose of description and should not be regarded as limiting.

The invention is herein described, by way of example only, with reference to the accompanying drawing. With specific reference now to the drawing in detail, it is stressed that the particulars shown are by way of example and for purposes of illustrative discussion of the preferred embodiments of the present invention only, and are presented in the cause



of providing what is believed to be the most useful and readily understood description of the principles and conceptual aspects of the invention. In this regard, no attempt is made to show structural details of the invention in more detail than is necessary for a fundamental understanding of the invention, the description taken with the drawing making apparent to those skilled in the art how the several forms of the invention may be embodied in practice.

The sole Figure shows polymer particle 1 encircled by a separate layer of RMR elastomer 2 in which glass particles 3 are dispersed. The elastomer material used in the new glass/polymer composites of the present invention has the capability of adhering the glass particles to the surface of the polymer particle. This elastomer may be, for example, an RMR liquid elastomer as described hereinabove. Use of an RMR elastomer facilitates curing with sulfur or peroxide.

According to preferred embodiments of the present invention, there is provided a glass/polymer composite material including an organic polymer and chalcogenide glass. Such composites are known to exhibit superior characteristics compared to non-composite polymer or glass taken individually. Improved characteristics include, but are not limited to, increased thermal stability, chemical stability, and enhanced fracture toughness.

According to preferred embodiments of the invention, the organic polymer constituent of the glass/polymer composite material has a low melting point, preferably below 200°C, more preferably below 180°C, more preferably below 160°C, more preferably below 140°C, more preferably below 120°C, and most preferably approximately 90 to 100°C.

According to preferred embodiments of the invention, the glass and polymer constituents of the glass/polymer composite material have the same or similar softening temperature points in the range of 90-200°C. It will be appreciated by those skilled in the art that the optimal softening temperature may be dependent upon the intended use of the composite material.

According to preferred embodiments of the invention, the glass and polymer constituents of the glass/polymer composite material have, alternately or additionally, the same or similar viscosity values in the range of 10-1000 PaS under extrusion conditions.

According to preferred embodiments of the invention, the glass/polymer contains low density polyethylene and chalcogenide glass

consisting of As - 15%, Sb - 3%, I - 28%, Se - 50%, Te - 4% ( $\text{As}_{15}\text{Sb}_3\text{I}_{28}\text{Se}_{50}\text{Te}_4$ ).

According to additional preferred embodiments of the invention, there is provided a glass/polymer composite material including: an organic polymer, a glass and a linear triblock copolymer elastomer. The elastomer may be, for example, an RMR elastomer, as described hereinabove. The elastomer functions to bond the glass to the polymer and may, alternately or additionally, aid in curing the composite.

According to preferred embodiments of the invention, the glass/polymer composite material includes glass selected from the group consisting of chalcogenide and alkali metal based glasses. The chalcogenide glasses have low softening temperatures and low viscosity, as detailed hereinabove, making them suitable for use in composites with polymers having similar physical properties. In addition, chalcogenide glasses have some unique optical properties, including but not limited to, transparency in the middle and far IR bands of the spectrum (in the interval: 2-20 mm), no dispersion in the in the middle and far IR bands of the spectrum, refractive indices of up to 2.5, and a photorefractive effect.

According to preferred embodiments of the invention, the glass/polymer composite material includes an elastomer which is a linear triblock copolymer having a narrow molecular weight distribution and having a center block which is a random ethene/butene backbone and terminal blocks which contain carbon-carbon double bond as taught by U.S. Patents 5,187,236 and 5,210,359.

According to preferred embodiments of the invention, the glass/polymer composite material includes low density polyethylene and chalcogenide glass consisting of As - 15%, Sb - 3%, I - 28%, Se - 50%, Te - 4% ( $\text{As}_{15}\text{Sb}_3\text{I}_{28}\text{Se}_{50}\text{Te}_4$ ) and elastomer selected from the group of linear triblock copolymers.

According to preferred embodiments of the invention, the glass/polymer composite material preferably includes polymer which constitutes from 90 to 99.5% by weight of the material, and glass which constitutes from 0.5 to 10% by weight of the material, more preferably 99% by weight polymer and 1% by weight glass.

According to preferred embodiments of the invention, the glass/polymer composite material is designed useful as a light filter, an optical window, a polarization layer, an adhesive and as a a light shaper/diffuser.

Additional preferred embodiments of the invention include a process for preparing a glass/polymer composite containing organic polymer particles and chalcogenide glass particles which includes the steps of: (a) mechanically milling of the glass particles to a size of  $< 100 \mu\text{m}$ ; (b) mixing the milled glass particles with the polymer particles; and (c) extruding the glass and polymer particles simultaneously in a melted form.

Further additional preferred embodiments of the invention include a process for producing a glass/polymer composite material including particles of an organic polymer, glass particles and linear triblock copolymer elastomer, which includes the steps of: (a) mechanically milling the glass particles to a final size of  $< 100 \mu\text{m}$ ; (b) homogeneously dispersing the milled glass particles in a solution of the elastomer in a solvent; (c) adding the polymer particles into the mixture of glass particles dispersed in the elastomer solution; (d) evaporating the solvent to form polymer particles coated by a layer of elastomer in which the glass particles are dispersed; and (e) extruding the glass and coated polymer particles simultaneously in a melted form.

According to preferred embodiments, the extrusion step of the process occurs under conditions of high shear stresses ( $10^5 - 10^6 \text{ Pa}$ ).

According to preferred embodiments, the process results in production of a film of 20-100  $\mu\text{m}$  thickness. This film, whether rigid or flexible, may be used to wrap or coat an object so that it acts as a barrier to liquids, water vapor and other gases.

The present invention successfully addresses the shortcomings of the presently known configurations by providing glass/plastic composites with low glass content and processes for forming same at relatively low temperatures and under conditions of high shear stress.

Additional objects, advantages, and novel features of the present invention will become apparent to one ordinarily skilled in the art upon examination of the following examples, which are not intended to be limiting. Additionally, each of the various embodiments and aspects of the present invention as delineated hereinabove and as claimed in the claims section below finds experimental support in the following examples.

### EXAMPLES

Reference is now made to the following examples, which together with the above descriptions, illustrate the invention in a non-limiting fashion.

### **Example 1**

#### ***Preparation of a glass /polymer composite and characterization of same***

A glass/polymer composite was formed using the chalcogenide glass having softening temperature point of 100° C., containing As - 15%, Sb - 3%, I - 28%, Se - 50%, Te - 4% ( $\text{As}_{15}\text{Sb}_3\text{I}_{28}\text{Se}_{50}\text{Te}_4$ ) and low density polyethylene IPETHENE-800 (produced by Carmel Olefins, Israel), having melt flow index (MFI) of 20g/10 min and density of 0.916 g/CC. The glass/polymer was prepared as follows: The chalcogenide glass was milled by the rotor beater mill to the final fineness <100  $\mu\text{m}$ . The powder size distribution was determined using optical microscope procedure. The size distribution was: 27% of particles have particle sizes greater than 30  $\mu\text{m}$ , 48% of particles have size between 10-30  $\mu\text{m}$ , 25% of particles have size less than 10  $\mu\text{m}$ .

The milled chalcogenide glass particles fraction was mixed (using dry mixer) with the granules of low density polyethylene at the proportion (w/w) of 99% polymer and 1% chalcogenide glass. The obtained mixture was extruded by a single-screw laboratory extruder (having screw diameter of 32 mm, ratio length of the extruder/diameter of the screw = 16). The parameters of extrusion included temperatures according to heating zones: 110° C, 120° C, 130° C, 135° C, speed of screw: 60 rotations/min and pressure (head) of 40 Atm.

Films of 20-100  $\mu\text{m}$  thickness were obtained by the direct screw extrusion procedure. The homogeneity of obtained films was determined using laser beam scanning (laser He-Ne, wavelength 0.63  $\mu\text{m}$ ) over the surface of the film and measuring optical transparency under the scanning procedure. It was established that within the experimental accuracy (2.5%) the transparency is constant under the scanning procedure.

The formed films were studied using a microscope, as well. It was observed that 100% of glass-particles have size less than 10  $\mu\text{m}$ .

### **Example 2**

#### ***Preparation of a glass /polymer composite with RMR liquid elastomer and characterization of same***

A glass/polymer composite was formed using the chalcogenide glass and low density polyethylene of example 1. The glass was milled as described in example 1.

RMR liquid elastomer (Mobil 10L) of approximately 10,000 molecular weight, was dissolved in cyclohexane in the proportion (v/v) of

5% RMR elastomer and 95% cyclohexane. The milled chalcogenide glass powder was dispersed in the obtained RMR elastomer solution. Granules of low density polyethylene were dipped and coated by the dispersion of chalcogenide glass in the RMR elastomer solution under dip-coating procedure. The cyclohexane was evaporated. Granules of low density polyethylene covered by the mix of RMR elastomer and chalcogenide glass particles were obtained. The Figure illustrates polymer particle 1 encircled by a separate layer 2 of RMR elastomer in which glass particles 3 are dispersed.

Preliminary homogenization of the blend was achieved. Blend mixture was extruded and 20-200  $\mu\text{m}$  thickness films were obtained as described in example 1. Films were studied with an optical microscope and it was established that 100% of particles of chalcogenide glass have size less 2  $\mu\text{m}$ .

Although the invention has been described in conjunction with specific embodiments thereof, it is evident that many alternatives, modifications and variations will be apparent to those skilled in the art. Accordingly, it is intended to embrace all such alternatives, modifications and variations that fall within the spirit and broad scope of the appended claims. All publications cited herein are incorporated by reference in their entirety. Citation or identification of any reference in this section or in any other section of this application shall not be construed as an admission that such reference is available as prior art to the present invention.

## WHAT IS CLAIMED IS:

1. A glass/polymer composite material comprising an organic polymer and chalcogenide glass.
2. The glass/polymer composite material of claim 1, wherein the organic polymer constituent has a low melting point.
3. The glass/polymer composite material of claim 1, wherein the glass and polymer constituents have the same or similar softening temperature points in the range of 90-200°C.
4. The glass/polymer composite material of claim 1, wherein the glass and polymer constituents have the same or similar softening temperature points in the range of 90-200°C and the same or similar viscosity values in the range of 10-1000 PaS under extrusion conditions.
5. The glass/polymer composite material of claim 1, comprising 90-99.5% by weight of polymer constituent and 0.5-10% by weight of glass constituent.
6. The glass/polymer composite material of claim 1, containing low density polyethylene and chalcogenide glass consisting of As - 15%, Sb - 3%, I - 28%, Se - 50%, Te - 4% ( $\text{As}_{15}\text{Sb}_3\text{I}_{28}\text{Se}_{50}\text{Te}_4$ ).
7. The glass/polymer composite material of claim 6, comprising about 99% by weight polymer and about 1% by weight glass.
8. The glass/polymer composite material of claim 1, wherein said composite is designed and is useful for an application selected from the group consisting a light filter, an optical window, a polarization layer, an adhesive and as a light shaper/diffuser.
9. A glass/polymer composite material comprising: an organic polymer, a glass and a linear triblock copolymer elastomer.
10. The glass/polymer composite material of claim 9, wherein said glass is selected from the group consisting of chalcogenide and alkali metal based glasses.

11. The glass/polymer composite material of claim 9, wherein said elastomer is a linear triblock copolymer having a narrow molecular weight distribution and having a center block which is a random ethene/butene backbone and terminal blocks which contain carbon-carbon double bond.

12. The glass/polymer composite material of claim 9, wherein said polymer constitutes from 90 to 99.5% by weight of the material, and wherein said glass constitutes from 0.5 to 10% by weight of the material.

13. The glass/polymer composite material of claim 9 including low density polyethylene and chalcogenide glass consisting of As - 15%, Sb - 3%, I - 28%, Se - 50%, Te - 4% ( $\text{As}_{15}\text{Sb}_3\text{I}_{28}\text{Se}_{50}\text{Te}_4$ ) and elastomer selected from the group of linear triblock copolymers.

14. The glass/polymer composite material of claim 9, wherein said composite is designed and is useful for an application selected from the group consisting a light filter, an optical window, a polarization layer, an adhesive and as a light shaper/diffuser.

15. The glass/polymer composite material of claim 13, containing 99% by weight polymer and 1% by weight glass.

16. A process for preparing a glass/polymer composite containing organic polymer particles and chalcogenide glass particles comprising the steps of:

- (a) mechanically milling of the glass particles to a size of  $< 100 \mu\text{m}$ ;
- (b) mixing the milled glass particles with the polymer particles; and
- (c) extruding the glass and polymer particles simultaneously in a melted form.

17. The process of claim 16, wherein the extrusion is made under conditions of high shear stresses ( $10^5 - 10^6 \text{ Pa}$ ).

18. A film of  $20-100 \mu\text{m}$  thickness produced by the process of claim 16.

19. The glass/polymer composite material produced by the process of claim 16, wherein said composite is designed and is useful for an application selected from the group consisting a light filter, an optical window, a polarization layer, an adhesive and as a light shaper/diffuser.

20. A process for producing a glass/polymer composite material including particles of an organic polymer, glass particles and linear triblock copolymer elastomer, comprising the steps of:

- (a) mechanically milling the glass particles to a final size of  $< 100 \mu\text{m}$ ;
- (b) homogeneously dispersing said milled glass particles in a solution of the elastomer in a solvent;
- (c) adding the polymer particles into the mixture of glass particles dispersed in said elastomer solution;
- (d) evaporating said solvent to form polymer particles coated by a layer of elastomer in which the glass particles are dispersed; and
- (e) extruding the glass and coated polymer particles simultaneously in a melted form.

21. The process of claim 20, wherein extrusion is made under conditions of high shear stresses ( $10^5 - 10^6 \text{ Pa}$ ).

22. A film of 20-100  $\mu\text{m}$  thickness produced by the process of claim 20.

23. The glass/polymer composite material produced by the process of claim 17, wherein said composite is designed and is useful for an application selected from the group consisting of a light filter, an optical window, a polarization layer, an adhesive and as a light shaper/diffuser.



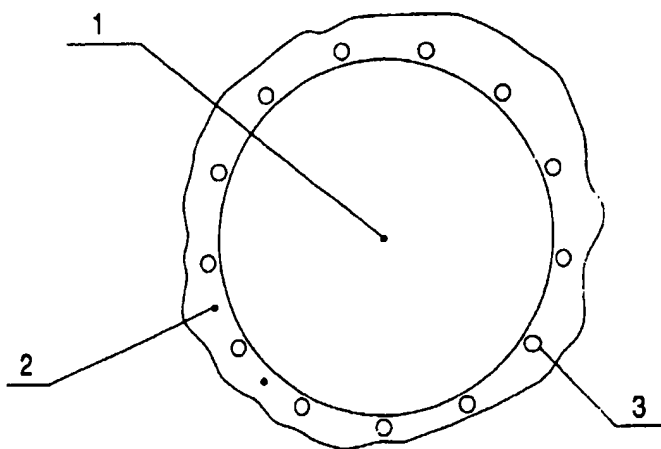


FIG.1

## INTERNATIONAL SEARCH REPORT

International application No.  
PCT/IL00/00054

<b>A. CLASSIFICATION OF SUBJECT MATTER</b> IPC(7) : C08K 03/32 US CL : 523/170; 524/8, 505 According to International Patent Classification (IPC) or to both national classification and IPC														
<b>B. FIELDS SEARCHED</b> Minimum documentation searched (classification system followed by classification symbols) U.S. : 523/170; 524/8, 505  Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched NONE  Electronic data base consulted during the international search (name of data base and, where practicable, search terms used) NONE														
<b>C. DOCUMENTS CONSIDERED TO BE RELEVANT</b>														
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.												
X — A	US 5,422,384 A (SAMUELS et al) 06 June 1995, see entire document.	1-5, 8, 18, 19, 22, 23 — 6, 7, 9-17, 20, 21												
A	US 4,965,783 A (SIOL et al) 23 October 1990, see entire document.	1-23												
<input type="checkbox"/> Further documents are listed in the continuation of Box C. <input type="checkbox"/> See patent family annex.														
<table border="0"><tr><td>* Special categories of cited documents:</td><td>"T" later documents published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention</td></tr><tr><td>"A" document defining the general state of the art which is not considered to be of particular relevance</td><td>"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone</td></tr><tr><td>"E" earlier document published on or after the international filing date</td><td>"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art</td></tr><tr><td>"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)</td><td>"Z" document member of the same patent family</td></tr><tr><td>"O" documents referring to an oral disclosure, use, exhibition or other means</td><td></td></tr><tr><td>"P" document published prior to the international filing date but later than the priority date claimed</td><td></td></tr></table>			* Special categories of cited documents:	"T" later documents published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention	"A" document defining the general state of the art which is not considered to be of particular relevance	"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone	"E" earlier document published on or after the international filing date	"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art	"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)	"Z" document member of the same patent family	"O" documents referring to an oral disclosure, use, exhibition or other means		"P" document published prior to the international filing date but later than the priority date claimed	
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Date of the actual completion of the international search 12 MAY 2000		Date of mailing of the international search report 06 JUL 2000												
Name and mailing address of the ISA/US Commissioner of Patents and Trademarks Box PCT Washington, D.C. 20231 Facsimile No. (703) 305-3230		Authorized officer BLAINE R. COPENHEAVER Telephone No. (703) 308-0661												